

Novel Isotope Effects on the Pairing Pseudogap in High- T_c Cuprates: Evidences for Polaronic Metal and Precursor BCS-Like Pairing of Large Polarons

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We have studied the novel isotope effects on the pairing pseudogap in underdoped and optimally doped cuprates within the large-polaron model and two non-standard BCS-like approaches. We have shown that in the intermediate-coupling regime the precursor pairing of large polarons occurs at a mean-field temperature $T^* > T_c$ and the near-absent, sizable and very large oxygen and copper isotope effects on T^* exist in cuprates with small and large Fermi surfaces. Our results for T^* , isotope shifts and exponents in slightly underdoped and optimally doped cuprates are in quantitative agreement with existing experiments and explain the discrepancy between various experiments.

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Superconductivity in ordinary and polaronic metals is the result of two distinct quantum phenomena (as argued in Refs.[1–5]): pairing of charge carriers at a characteristic temperature T^* and condensation of bound pairs into a superfluid state at the superconducting (SC) transition temperature T_c . In conventional superconductors, pairing of electrons and superfluidity of Cooper pairs occur simultaneously at $T^* = T_c$. In high- T_c cuprates, however, the pairing of charge carriers may occur at a higher temperature T^* than the T_c at which the pre-formed Cooper pairs condense into a superfluid (or SC) state [1–3]. As a consequence, below T^* a pseudogap (PG) appears in cuprates, as seen in many experiments (NMR, ARPES, optical conductivity, specific heat, etc.) (see Refs.[6–8] for a review). Most of the theories proposed for the high- T_c cuprates are based on different electronic mechanisms of pairing and ignore effects arising from the electron-phonon interactions. However, a number of experiments revealing the peculiar isotope effects on T_c , T^* and other physical quantities in cuprates [9–15] have shown that the electron-phonon interactions play a major role in these materials. In particular, some experiments showed that the oxygen and copper isotope effects on T^* in Y- and La- based cuprates are absent or very small [9, 14] and sizable [9, 13, 14]. While other experiments revealed the presence of large negative oxygen and copper isotope effects on T^* in Ho-based cuprates [11, 13]. The origin of the novel isotope effects on T^* in cuprates has especially been the subject of controversy [9–11]. Although some theories [16–18] were used to describe the oxygen isotope effect on T^* in high- T_c cuprates, they cannot give an answer to the puzzling question why such an isotope effect is sizable or very large in some cuprates and absent in others. Also, there is no explanation of the observed copper isotope effect on T^* in cuprates.

In this Letter, we address the above issues of the isotope effects on the pairing PG in the cuprates by proposing two BCS-based approaches extended to the

intermediate-coupling regime, which allow to describe the precursor pairing of large polarons above T_c in underdoped and optimally doped cuprates with small and large Fermi surfaces. We derive new expressions for the mean-field pairing temperature T^* and the isotope exponents α_{T^*} . Remarkably, our results for T^* , isotope shifts and exponents in slightly underdoped and optimally doped cuprates are in quantitative agreement with existing experiments and resolve the controversy between various experiments. We also make quantitative predictions for the isotope effects on T^* in the deeply underdoped cuprates.

In conventional superconductors the mass of free electrons is independent of the ionic mass M . In contrast, the charge carriers in polar materials are self-trapped and form large polarons (quasiparticles dressed by lattice distortions) [19]. In the polaronic model, the mass of polarons m_p depends on the longitudinal-optical (LO) phonon-frequency ω_{LO} which in turn depends on M . The observed effective mass of charge carriers in cuprates is about $2m_e$ (where m_e is the free electron mass) [20] and the relatively small binding energies $E_p = 0.06\text{eV}$ and $E_p = 0.12\text{eV}$ of the polaron are observed in optimally doped and underdoped cuprates, respectively [21]. These and other experiments [20, 22] prove that the charge carriers in cuprates are large polarons.

Here we consider the real physical situations in cuprates, namely, the cases of small and large Fermi surfaces. In slightly underdoped and optimally doped cuprates with large Fermi surfaces, the new situation arises when the polaronic effect exists and the attractive interaction mechanism (e.g., due to exchange of static and dynamic phonons) between the carriers operating in the energy range $\{-(E_p + \hbar\omega_{LO}), (E_p + \hbar\omega_{LO})\}$ is more effective than in the simple BCS picture. The energies ε of polarons are measured from the polaronic Fermi energy ε_F . It was argued [1–3] (see also Ref. [23]) that the extension of the BCS theory to the intermediate coupling regime describes the precursor pairing of carriers above T_c . At $\varepsilon_F > E_p + \hbar\omega_{LO} \gg k_B T^*$, using a similar BCS-based approach, we obtain a new and more general

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expression for the mean-field pairing temperature T^* :

$$k_B T^* = 1.14(E_p + \hbar\omega_{LO}) \exp[-1/\lambda^*], \quad (1)$$

where $\lambda^* = N_p(\varepsilon_F)\tilde{V}_p$ is the BCS-like coupling constant, $N_p(\varepsilon_F)$ is the polaronic density of states (DOS), $\tilde{V}_p = V_{ph} - V_c$ is the effective polaron-polaron interaction potential, which has both the attractive electron-phonon interaction part V_{ph} and the screened repulsive Coulomb interaction part $\tilde{V}_c = V_c/[1 + N_p(\varepsilon_F)V_c \ln(\zeta_c/(E_p + \hbar\omega_{LO}))]$, ζ_c is the cut-off for the Coulomb interaction V_c , $\zeta_c > E_p + \hbar\omega_{LO}$. The usual BCS approximation as the particular case is recovered in the weak coupling regime (i.e. in the absence of polaronic effects, $E_p = 0$) and the prefactor in Eq.(1) is replaced by Debye energy $\hbar\omega_D$. Here we show that the polaronic effects change significantly the simple BCS picture and are responsible for the novel isotope effects on T^* . In the large polaron theory, m_p , E_p and $\varepsilon_F = \hbar^2(3\pi^2 n)^{2/3}/2m_p$ (where n is the concentration of large polarons) depend on the Fröhlich-type electron-phonon coupling constant α_F which in turn depends on the masses $M(= M_O$ or $M_{Cu})$ and $M'(= M_{Cu}$ or $M_O)$ of the oxygen O and copper Cu atoms in cuprates:

$$\alpha_F = \frac{e^2}{2\hbar\omega_{LO}} \left[\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right] \left(\frac{2m^*\omega_{LO}}{\hbar} \right)^{1/2}, \quad (2)$$

where $\omega_{LO} \simeq \left(2\beta \left(\frac{1}{M} + \frac{1}{M'} \right) \right)^{1/2}$, β is a force constant of the lattice, ε_∞ and ε_0 are the high frequency and static dielectric constants, respectively, m^* is the effective mass of a carrier in the absence of the electron-phonon interaction. In the intermediate electron-phonon coupling regime the mass and binding energy of a large polaron are given by $m_p = m^*(1 + \alpha_F/6)$ and $E_p = \alpha_F \hbar\omega_{LO}$ [19]

The polaronic DOS can be approximated in a simple form

$$N_p(\varepsilon_F) = \begin{cases} 1/\varepsilon_F & \text{for } 0 < \varepsilon \leq \varepsilon_F \\ 0 & \text{otherwise.} \end{cases} \quad (3)$$

From Eq.(1), the exponent of the isotope effect on T^* is defined as

$$\alpha_{T^*} = -\frac{d \ln T^*}{d \ln M}. \quad (4)$$

Using the above expressions for m_p , E_p and $N_p(\varepsilon_F)$, the Eqs. (1) and (4) can be written as

$$k_B T^* = 1.14 A \mu^{-1/4} \left(1 + a \mu^{-1/4} \right) \exp[-1/\lambda^*(\mu)] \quad (5)$$

and

$$\alpha_{T^*} = \frac{1}{4(1 + M/M')} \left\{ 1 + \frac{a \mu^{-1/4}}{1 + a \mu^{-1/4}} - \frac{1}{(\lambda^*(\mu))^2} \left(\lambda_{ph} b \mu^{1/4} - \frac{\lambda_c b \mu^{1/4}}{U_c(\mu)} \right. \right. \\ \left. \left. + \frac{\lambda_c^2(1 + b \mu^{1/4})}{U_c^2(\mu)} \left[b \mu^{1/4} \ln B_c(\mu) + (1 + b \mu^{1/4}) \left(1 + \frac{a \mu^{-1/4}}{1 + a \mu^{-1/4}} \right) \right] \right) \right\}, \quad (6)$$

where $\lambda^*(\mu) = \lambda_{ph}(1 + b \mu^{1/4}) - \lambda_c(1 + b \mu^{1/4})/U_c(\mu)$, $U_c(\mu) = 1 + \lambda_c(1 + b \mu^{1/4}) \ln B_c(\mu)$, $\lambda_{ph} = [2m^*/\hbar^2(3\pi^2 n)^{2/3}] V_{ph}$, $\lambda_c = [2m^*/\hbar^2(3\pi^2 n)^{2/3}] V_c$, $B_c(\mu) = \zeta_c/A \mu^{-1/4} (1 + a \mu^{-1/4})$, $A = \frac{e^2}{\tilde{\varepsilon}} \sqrt{\frac{m^*}{2\hbar}} (2\beta)^{1/4}$, $a = \hbar \tilde{\varepsilon} \sqrt{\frac{2\hbar}{m^*}} (2\beta)^{1/4} / e^2$, $b = 1/6a$, $\tilde{\varepsilon} = \varepsilon_\infty / (1 - \varepsilon_\infty/\varepsilon_0)$, $\mu = MM'/(M + M')$ is the reduced mass of ions.

Equations (5) and (6) allow us to calculate the PG temperature T^* and the exponents $\alpha_{T^*}^O$ and $\alpha_{T^*}^{Cu}$ of the oxygen and copper isotope effects on T^* . In our numerical calculations, we take $m^* \simeq m_e$ [20], $\varepsilon_\infty = 3 - 5$ [20, 24], $\varepsilon_0 = 22 - 30$ [20, 24] and $\hbar\omega_{LO} = 0.04 - 0.07 eV$ [7, 20], typical values for the cuprates. Then the values of $\tilde{\varepsilon}$ and α_F are $\tilde{\varepsilon} \simeq 3.33 - 6.47$ and $\alpha_F = 2.15 - 5.54$ (which correspond to the intermediate electron-phonon coupling regime). Notice that in discussing the experimental data for isotope effects on T^* in cuprates we have taken the fact that the physical situations (doping levels, dielectric constants, T^*) in various experiments are

rather different. The magnitude of β is kept at the value estimated for the oxygen and copper unsubstituted compound using a given value of $\hbar\omega_{LO}$. Since the quantity ζ_c is of the order of ε_F , the logarithm $\ln B_c(\mu)$ will be small, so that the Coulomb pseudopotential \tilde{V}_c is of the order of bare Coulomb potential V_c . The results of numerical calculations of T^* and α_{T^*} according to Eqs.(5) and (6) at different values of $\hbar\omega_{LO}$, $\tilde{\varepsilon}$, n , λ_{ph} and λ_c are shown in Figs.1 - 3.

Our results provide a consistent picture of the existence of crossover temperature T^* above T_c and peculiar isotope effects on T^* in cuprates. They explain why the small positive (see Fig.1) and very large negative (see Figs.2 and 3) oxygen isotope effects and the large negative and near-absent copper isotope effects on T^* are observed in various experiments. The obtained T^* is plotted in the insets of Figs. 1 - 3 as a function of $\tilde{\varepsilon}$ and n for different values of λ_{ph} and λ_c . The values of λ^* varies from 0.3 to 0.5 and T^* increases with decreasing n . We have verified that T^* decreases with in-

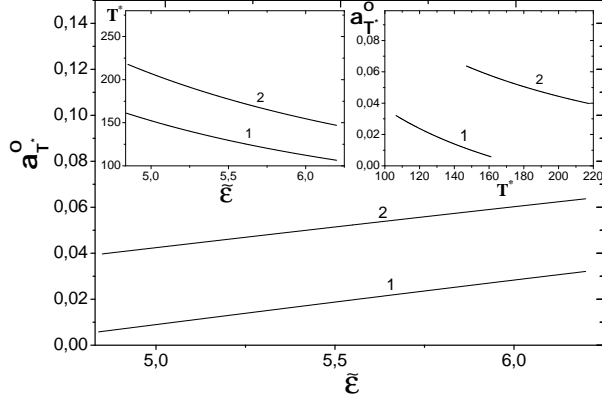


FIG. 1: Variation of α_{T*}^O as a function of $\tilde{\epsilon}$ for two sets of parameters: (1) $\lambda_{ph} = 0.297$, $\lambda_c = 0.077$, $n = 0.92 \cdot 10^{21} \text{cm}^{-3}$ and (2) $\lambda_{ph} = 0.313$, $\lambda_c = 0.067$, $n = 0.94 \cdot 10^{21} \text{cm}^{-3}$. The insets show the dependences $T^*(\tilde{\epsilon})$ and $\alpha_{T*}^O(\tilde{\epsilon})$ for the same values of λ_{ph} , λ_c , n .

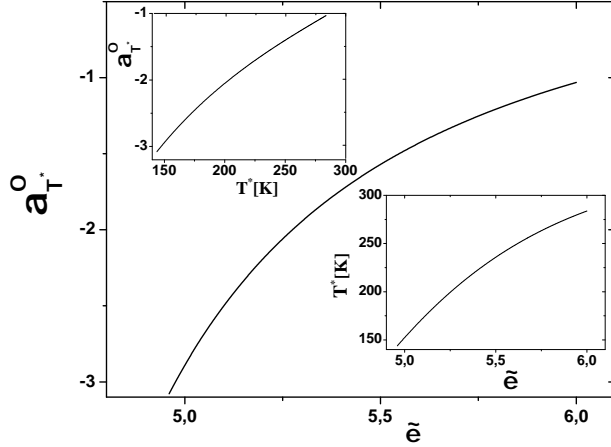


FIG. 2: The dependence of α_{T*}^O on $\tilde{\epsilon}$ for $\lambda_{ph} = 0.975$, $\lambda_c = 0.820$ and $n = 0.88 \cdot 10^{21} \text{cm}^{-3}$. The insets show the dependences $T^*(\tilde{\epsilon})$ and $\alpha_{T*}^O(\tilde{\epsilon})$ for the same values of λ_{ph} , λ_c , n .

creasing $\tilde{\epsilon}$ for $\lambda_{ph} < 0.35$ and $\lambda_c < 0.1$ (Fig.1). In contrast, T^* increase with increasing $\tilde{\epsilon}$ for $\lambda_{ph} > 0.4$ and $\lambda_c > 0.25$ (Fig.2). The existing experimental data on T^* and α_{T*}^O could be fitted with an excellent agreement using Eqs. (5) and (6), and adjusting the parameters $\hbar\omega_{LO}$, $\tilde{\epsilon}$, n , λ_{ph} and λ_c for each cuprate superconductor. If we choose $\hbar\omega_{LO} = 0.05 \text{eV}$, $n = 0.92 \cdot 10^{21} \text{cm}^{-3}$, $\tilde{\epsilon} = 4.841 - 5.045$, $\lambda_{ph} \simeq 0.297$ and $\lambda_c \simeq 0.077$, we see that $T^* = 150 - 161 \text{K}$ and α_{T*}^O is very small (i.e., $\alpha_{T*}^O = (0.0058 - 0.0098) < 0.01$), which are consistent with the experimental data of Ref. [9]. Further, using other sets of parameters $n = 0.94 \cdot 10^{21} \text{cm}^{-3}$, $\tilde{\epsilon} = 5.904 - 6.119$, $\lambda_{ph} = 0.311 - 0.313$ and $\lambda_c = 0.067 - 0.070$, we obtain $T^* \approx 150 \text{K}$ and $\alpha_{T*}^O \simeq 0.0525 - 0.0623$, which are in good agreement with the measured values: $T^* = 150 \text{K}$ and

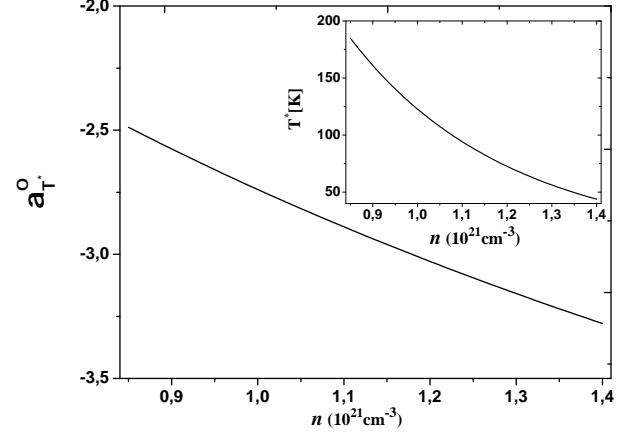


FIG. 3: The doping dependence of α_{T*}^O (main figure) and T^* (inset) for $\lambda_{ph} = 0.975$, $\lambda_c = 0.820$ and $\tilde{\epsilon} = 5.088$.

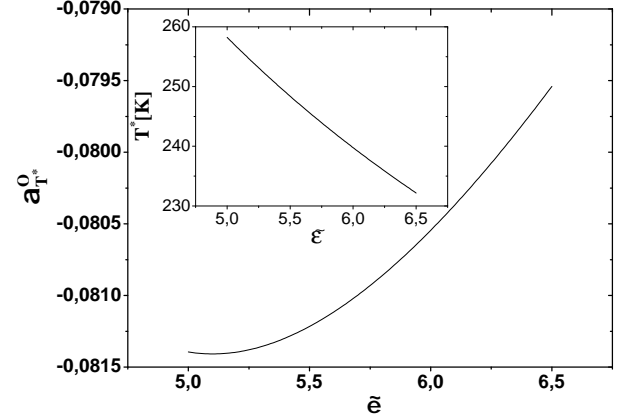


FIG. 4: Variation of α_{T*}^O (main figure) and T^* (inset) as a function of $\tilde{\epsilon}$ for $\lambda_{ph} - \lambda_c = 0.3$ and $n = 0.6 \cdot 10^{21} \text{cm}^{-3}$.

$\alpha_{T*}^O = 0.052 - 0.061$ for $\text{YBa}_2\text{Cu}_4\text{O}_8$ (with $T_c = 81 \text{K}$) [10]. Figure 1 illustrates the predicted behaviors of α_{T*}^O and T^* as a function of $\tilde{\epsilon}$ for $\lambda_{ph} < 0.4$ and $\lambda_c < 0.1$. We see that α_{T*}^O decreases slowly with decreasing $\tilde{\epsilon}$. Relatively strong electron-phonon and Coulomb interactions (i.e., $\lambda_{ph} > 0.5$ and $\lambda_c > 0.5$) change the picture significantly and cause α_{T*}^O to decrease rapidly with decreasing $\tilde{\epsilon}$. In this case the value of α_{T*}^O is negative and becomes very large negative with decreasing $\tilde{\epsilon}$ or T^* . The pictures shown in Figs.2 and 3 are likely realized in some cuprates (which exhibit a large negative isotope exponent α_{T*}^O) and explain another important puzzle of the cuprates [11]: the huge oxygen-isotope effect on T^* observed in $\text{HoBa}_2\text{Cu}_4\text{O}_8$, whose characteristic PG temperature T^* increases significantly upon replacing ^{16}O by ^{18}O . Indeed, with fitting parameters, $n = 0.88 \cdot 10^{21} \text{cm}^{-3}$, $\tilde{\epsilon} = 5.088$, $\lambda_{ph} = 0.975$ and $\lambda_c = 0.82$, one can ex-

plain the observed experimental data of Ref. [11]. In this case, we obtain $T^*(^{16}\text{O}) \simeq 170.2\text{K}$, $T^*(^{18}\text{O}) \simeq 220.2\text{K}$, $\Delta T_O^* = T^*(^{18}\text{O}) - T^*(^{16}\text{O}) \simeq 50\text{K}$ and $\alpha_{T^*}^O \simeq -2.54$, which are in remarkably good agreement with the experimental data $T^*(^{16}\text{O}) \simeq 170\text{K}$, $T^*(^{18}\text{O}) \simeq 220\text{K}$, $\Delta T_O^* \simeq 50\text{K}$ and $\alpha_{T^*}^O \simeq -2.2 \pm 0.6$ [11]. The above predicted behaviors of T^* and $\alpha_{T^*}^O$ could be checked experimentally in other slightly underdoped and optimally doped cuprates. We have also performed similar calculations for the copper isotope effect on T^* in slightly underdoped $\text{HoBa}_2\text{Cu}_4\text{O}_8$ (where the electron-phonon and Coulomb interactions seem to be much stronger than in $\text{YBa}_2\text{Cu}_4\text{O}_8$), and for the oxygen and copper isotope effects on T^* in optimally doped $\text{La}_{1.81}\text{Ho}_{0.04}\text{Sr}_{0.15}\text{CuO}_4$. In order to obtain the values of $T^* \simeq 160$ and $\simeq 185\text{K}$ observed accordingly in copper unsubstituted and substituted $\text{HoBa}_2\text{Cu}_4\text{O}_8$, we took $\hbar\omega_{LO} = 0.05\text{eV}$, $n = 0.9 \cdot 10^{21}\text{cm}^{-3}$, $\tilde{\varepsilon} = 4.937$, $\lambda_{ph} = 2.196$, $\lambda_c = 2.064$. Then we found $T^*(^{63}\text{Cu}) \simeq 160\text{K}$, $T^*(^{65}\text{Cu}) \simeq 184.7\text{K}$, $\Delta T_{Cu}^* = T^*(^{65}\text{Cu}) - T^*(^{63}\text{Cu}) \approx 25\text{K}$ and $\alpha_{T^*}^{Cu} \simeq -4.9$ in accordance with experimental findings $T^*(^{63}\text{Cu}) \approx 160\text{K}$, $T^*(^{65}\text{Cu}) \approx 185\text{K}$ and $\alpha_{T^*}^{Cu} \simeq -4.9$ [13]. In the orthorhombic $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ the optimally doped level ($x \simeq 0.15$) corresponds to the value $n = 0.8 \cdot 10^{21}\text{cm}^{-3}$. By taking $\hbar\omega_{LO} = 0.045\text{eV}$, $\tilde{\varepsilon} = 5.681$, $\lambda_{ph} = 0.578$ and $\lambda_c = 0.463$ for $\text{La}_{1.81}\text{Ho}_{0.04}\text{Sr}_{0.15}\text{CuO}_4$, we obtained $T^*(^{16}\text{O}) = T^*(^{63}\text{Cu}) \simeq 60\text{K}$, $T^*(^{18}\text{O}) \simeq 69.3\text{K}$, $\Delta T_O^* \simeq 9.3\text{K}$, $\alpha_{T^*}^O \simeq -1.3$, $T^*(^{65}\text{Cu}) \simeq 60.7\text{K}$; $\Delta T_{Cu}^* \simeq 0.7\text{K}$, which agree with the experimental data of Ref. [14].

The situation is different for the deeply underdoped cuprate with a small Fermi surface. In this case, the polaronic effect is strong enough and the condition $\varepsilon_F < E_p$ is satisfied. Therefore, $E_p + \hbar\omega_{LO}$ and λ^* in Eq.(1) would be replaced by ε_F and $\lambda^* = (V_{ph} - V_c)N_p(\varepsilon_F)$, respectively. It is easy to verify numerically that, for $\varepsilon_F/k_B T^* \gtrsim 3.7$ (or $\lambda^* \lesssim 0.7$), the PG temperature T^* can be determined from the BCS-like relation

$$k_B T^* \simeq 1.14 \varepsilon_F \exp(-1/\lambda^*(\mu)), \quad (7)$$

where $\lambda^*(\mu) = (\lambda_{ph} - \lambda_c)(1 + b\mu^{1/4})$.

Using Eqs. (4) and (7), we obtain

$$\alpha_{T^*} = \frac{b\mu^{\frac{1}{4}}}{4(1 + \frac{M}{M'}) (1 + b\mu^{1/4})} \left[1 - \frac{1}{\lambda^*(\mu)} \right]. \quad (8)$$

The dependences $\alpha_{T^*}^O(\tilde{\varepsilon})$ and $T^*(\tilde{\varepsilon})$ for $\hbar\omega_{LO} = 0.05\text{eV}$, $\lambda_{ph} - \lambda_c = 0.3$ and $n = 0.6 \cdot 10^{21}\text{cm}^{-3}$ are presented in Fig.4. As seen from Fig.4, the $\alpha_{T^*}^O$ is rather small and negative. The $\alpha_{T^*}^{Cu}$ is also negative and nearly four times smaller than $\alpha_{T^*}^O$.

In conclusion, we have developed two new BCS-based approaches extended to the intermediate coupling regime to describe the precursor pairing of large polarons above T_c and the peculiar isotope effects on the PG temperature T^* in high- T_c cuprates with small and large Fermi surfaces. Our results show that the oxygen isotope effect on T^* in optimally doped cuprates with moderate electron-phonon coupling strength ($\lambda_{ph} < 0.4$) and weak Coulomb repulsion ($\lambda_c < 0.1$) is small positive. At $\lambda_{ph} > 0.5$ and $\lambda_c < 0.5$ the negative oxygen and copper isotope effects on T^* in the same compound (e.g., $\text{La}_{1.81}\text{Ho}_{0.04}\text{Sr}_{0.15}\text{CuO}_4$) is large and negligible, respectively. While the very large negative oxygen and copper isotope effects on T^* exist in some cuprates with large Fermi surfaces at $\lambda_{ph} < 1$, $\lambda_c \lesssim 0.8$ and $\lambda_{ph} > \lambda_c > 2$, respectively. The existing experimental data and discrepancies between experiments measuring the isotope effects on T^* in various cuprates are consistently explained by the proposed general BCS-like pairing model. We predict that the isotope effects on T^* in the deeply underdoped cuprates are sizable and negative.

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